

DOCKET NO: 215150US0

**IN THE UNITED STATES PATENT & TRADEMARK OFFICE**

IN RE APPLICATION OF :  
GOTTLIEB-GEORG LINDNER, ET AL. : EXAMINER: NGUYEN  
SERIAL NO: 10/079,479 :  
FILED: FEBRUARY 22, 2002 : GROUP ART UNIT: 1793  
FOR: SILICA BY PRECIPITATION AT  
CONSTANT ALKALI NUMBER, AND ITS USE

**APPEAL BRIEF**

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

This is an appeal from the Examiner's Final Rejection dated September 22, 2008, of Claims 1, 4, 5, 10-12, 14-19 and 22-28. A Notice of Appeal was filed on December 22, 2008.

**I. REAL PARTY IN INTEREST**

The real party in interest is Evonik Degussa GmbH of Germany, by virtue of the assignment recorded January December 9, 2008, at Reel/Frame 021942/0139.

**II. RELATED APPEALS AND INTERFERENCES**

Appellants, Appellants' legal representative and their assignee are not aware of any other appeals or interferences which will directly affect or be directly affected by or having a bearing on the Board's decision in this appeal.

### **III. STATUS OF CLAIMS**

The appealed claims are Claims 1, 4, 5, 10-12, 14-19 and 22-28. Claims 1, 4, 5, 10-12, 14-19 and 22-28 stand rejected.

The status of Claims 1, 4, 5, 10, 11, 14-18 and 22-28 is “previously presented”. The status of Claims 12 and 19 is “original”. The status of Claims 2, 3, 6-9, 13, 20-21 is “canceled”.

### **IV. STATUS OF AMENDMENTS**

No Amendment under 37 C.F.R. §1.116 was filed.

### **V. SUMMARY OF CLAIMED SUBJECT MATTER**

As claimed in **Claim 1**, the present application relates to a precipitated silica having the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g;
DBP/choline chloride absorption	less than 1.07; and
Sears number	greater than 25 ml/5g.

See for example, page 4, lines 1-13 and 20-24 of the specification.

As set forth in **Claim 10**, the present invention relates to a process for preparing precipitated silica, comprising:

simultaneously metering into an aqueous silicate solution more aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,

optionally filtering the acidified mixture to obtain a filtered precipitated silica,

optionally drying the filtered precipitated silica,

wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15, and

wherein said silica has the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g;
DBP/choline chloride absorption	less than 1.07;
Sears number	greater than 25 ml/5g.

See for example, page 2, lines 7-16, page 3, lines 21-26, page 4, lines 1-13 and 20-24, the general description of the Examples at page 5 and Examples 5 and 6 of the specification.

As set forth in **Claim 17**, the present invention relates to a process for preparing precipitated silica, comprising:

simultaneously metering into a vessel an aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,

optionally filtering the acidified mixture to obtain a filtered precipitated silica,

optionally drying the filtered precipitated silica,

wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15, and

wherein said silica has the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g; and
DBP/choline chloride absorption	less than 1.07;
Sears number	greater than 25 ml/5g.

See for example, page 2, lines 7-16, page 3, lines 21-26, page 4, lines 1-13 and 20-24, the general description of the Examples at page 5 and Examples 5 and 6 of the specification.

The specification states at page 1, lines 13 to 26, at page 1, line 29-33 and at page 2, lines 2-4 states the following:

“Precipitated silicas as carrier materials, particularly for vitamin E acetate or choline chloride have long been known. For example, EP 0 937 755 describes how a precipitated silica is prepared by a pH-controlled precipitation reaction and then

spray-dried. Precipitated silica prepared in this way is particularly suitable for use for adsorbing liquid active substances such as choline chloride solution or vitamin E, for example. DE 198 60 441 discloses how an active substance adsorbate may be prepared from a precipitated silica and an active substance by spraying or injecting a silica suspension together with one or more active substances into a fluidized bed generated using hot air. It is likewise possible to use hydrophobic precipitated silicas for these purposes, as described in DE 198 25 687.

In the context of their use as carriers, the following properties of silicas are important: adsorption capacity, good sorption kinetics, and low fine dust fraction. Owing to heightened safety requirements and the need to prepare adsorbates with ever higher concentrations, there is therefore a demand for carrier silicas which have a very low fines fraction with an adsorptiveness which is heightened at the same time.”

“The known silicas generally do not possess pronounced sorption characteristics for polar compounds. Since silicas are frequently used as carrier material for polar compounds such as choline chloride, propionic acid or formic acid, for example, it is one object of the present invention to provide a silica and a silicate which possesses particularly good sorbency for polar compounds.”

“It has surprisingly been found that by preparing precipitated silica and silicate at a constant alkali number, products can be obtained which have good sorption characteristics for polar compounds.”

## **VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

(A) Claims 1, 4-5, 10-12, 14-19 and 22-28 stand rejected as being obvious under 35 U.S.C. §103 (a) over EP ‘755 (EP 0 937 755) in view of Turk et al (US 4,001, 379).

## **VII. ARGUMENT**

Ground (A)

Claims 1, 4-5, 10-12, 14-19 and 22-28 stand rejected as being obvious under 35 U.S.C. §103 (a) over EP ‘755 in view of Turk et al. That rejection is untenable and should not be sustained.

EP ‘755 and Türk, alone or in combination, fail to disclose or suggest a precipitated silica having the following **physicochemical characteristics** as claimed in **Claims 1, 10 and 17.**

<b>BET surface area</b>	<b>from 50 to 700 m<sup>2</sup>/g;</b>
<b>DBP absorption</b>	<b>from 100 to 450 g/100 g;</b>
<b>Choline chloride absorption</b>	<b>from 150 to 400 g/100 g (75% absorption by weight aqueous solution);</b>
<b>CTAB surface area</b>	<b>from 50 to 350 m<sup>2</sup>/g;</b>
<b>DBP/choline chloride absorption</b>	<b>less than 1.07; and</b>
<b>Sears number</b>	<b><u>greater than 25 ml/5g.</u></b>

Further, EP '755 and Türk, alone or in combination, fail to disclose or suggest a process as claimed in Claim 10 for preparing precipitated silica having the combination of properties as claimed in Claim 10, comprising:

**simultaneously metering into an aqueous silicate solution more aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,**

**wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15.**

Moreover, EP '755 and Türk, alone or in combination, fail to disclose or suggest a process as claimed in Claim 17 for preparing precipitated silica having the combination of properties as claimed in Claim 17, comprising:

**simultaneously metering into a vessel an aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,**

**wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15.**

Regarding Claims 10 and 17, the specific process steps are not disclosed or suggested in EP '755 and Türk, alone or in combination.

Regarding EP '755, the Examiner has argued in the Office Action of September 22, 2008, “since the product of EP '755 has all the same properties and being made by the same process … the Sears number of the product of EP '755 would inherently be the same as that of the claimed product. See page 3, 2<sup>nd</sup> full paragraph of the Office Action of September 22, 2008. However, as discussed below, the process of EP'755 is NOT the same as the claimed process and thus, the Sears number cannot be inherent. The methods for preparing the silicas of EP '755 are so-called “pH precipitation methods,” in which the pH is kept constant during the precipitation (see, for example, Examples 1 and 2 of EP '755). The method of the present invention is a so-called “AN precipitation method,” in other words the alkali number (AN) is kept constant. It was already pointed out in the specification of the present application that pH precipitation methods are something different, and lead to different precipitated silica than AN precipitation methods. See page 3, lines 6-20 of the specification which states:

“There are known preparation processes for precipitated silicas in the course of which a constant pH is maintained. A precipitation reaction at constant alkali number, on the other hand, means that the concentration of freely available alkali ions (e.g., sodium ions) is kept constant.

As a result of the acid-base reactions during the precipitation of waterglass with sulfuric acid, sodium ions are on the one hand released in the form of sodium sulfate; on the other hand, sodium ions are incorporated into the silicate agglomerates which form.

Since these two reactions proceed independently of one another kinetically, the course of precipitations at constant pH is different than that of precipitations conducted in accordance with the invention.

In the case of a precipitation reaction at constant alkali number, the pH changes analogously: for example, at a constant alkali number of 30, the pH falls from about 10.35 to

levels between 8 and 10, depending on the duration of the precipitation reaction (simultaneous addition of alkaline silicate solution and acid). The longer such a precipitation reaction lasts, the lower the pH at the end of the reaction is. The intercalation of sodium ions into the silica structure is probably responsible for this."

Further, *In re Oelrich*, 666 F.2d 578, 581 (CCPA 1981), [instructs,] the mere fact that a certain thing may result from a given set of circumstances is not sufficient to prove inherency. Inherency may not be established by probabilities or possibilities. Something that is inherent must inevitably be the result each and every time.

The fact that a certain result or characteristic may occur or may be present in the prior art is not sufficient to establish the inherency of that result or characteristic. *In re Rijckaert*, 9 F.3d 1531, 1534 (Fed. Cir. 1993). "To establish inherency, the extrinsic evidence 'must make clear that the missing descriptive matter is necessarily present in the thing described in the reference, and that it would be so recognized by persons of ordinary skill. Inherency, however, may not be established by probabilities or possibilities. The mere fact that a certain thing may result from a given set of circumstances is not sufficient.'" *In re Robertson*, 169 F.3d 743, 745 (Fed. Cir. 1999)(citations omitted). An invitation to experiment is not an inherent disclosure. *Metabolite Labs., Inc. v. Lab. Corp. of Am. Holdings*, 370 F.3d 1354, 1367 (Fed. Cir. 2004).

Finally, *Ex parte Levy*, 17 USPQ2d 1461, 1464 (Bd. Pat. App. & Inter. 1990), instructs:

In relying upon the theory of inherency, the examiner must provide a basis in fact and/or technical reasoning to reasonably support the determination that the allegedly inherent characteristic necessarily flows from the teachings of the applied prior art.

In this case, the Examiner has not provided a basis in fact and/or technical reasoning to support a finding that the allegedly inherent characteristic (Sears number) necessarily

flows from EP '755. Therefore, the Examiner's rejection of Applicant's claims based on the Examiner's finding of inherency cannot stand and should be reversed.

Further, the Examiner refers to Examples 3 of EP'755. However, the method according to Example 3 of EP '755 nevertheless differs decisively from that of the present invention, as the following table shows:

Method parameters	EP '755, Example 3	Invention, Examples 1-8
Precipitation time (min)	90	40-65
Precipitation temperature (°C)	91-93	85
Alkali number	7	20-40
Solid content of the suspension (g/l)	72	80-105

As can be seen, numerous significant parameters of the method of the present invention are clearly different. Thus, the precipitation time is not even half as long as in EP '755, in some instances. The alkali number is 3-6 times greater. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

The Examiner has previously argued that a Sears number of greater than 25 ml/5g as claimed is obvious and expected in view of the Sears number of  $22.6 \pm 0.25$  ml/5g of Product C Versuch 13 of EP '755 and in view of  $19.9 \pm 0.25$  ml/5g of Product A Versuch 6 of EP '755. However, there is a difference of at least 10 % between the claimed Sears number and the Sears number of EP '755. The margin of error is very small ( $\pm 0.25$  ml/5g). This was already discussed in the Rule 132 Declaration of Dr. Ralph Rausch, signed on April 18, 2007

(Exhibit A) and the Rule 132 Declaration of Dr. Görl signed October 19, 2007 (Exhibit B).

See also the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

The increase of the Sears number by at least 10% compared to EP ‘755 is significant because it is necessary to achieve a sufficient affinity between the silica of the invention and hydrophilic liquids. Without such affinity the desired DBP/CC ratio of below 1.07 cannot be reached. The inventors have found that for a given set of other physico-chemical properties of a silica, a borderline exists for the Sears number which has to be exceeded. Therefore an increase of 10 % in Sears number is significant. See item 13 in the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C). Applicants wish to draw the Examiners' attention to each of items 3 to 13 of the Rule 132 Declaration signed May 13, 2008. Each item discussed in each of the three Declarations presents evidence on the record. The Examiner is requested to consider each item carefully. The various items are again discussed below.

The difference of at least 10 % between the claimed Sears number and the Sears number of EP ‘755 is not obvious. It is very difficult to increase the Sears number and at the same time keep all other parameter as claimed in the ranges as claimed. The Sear number is rather complex in its emergence: factors like temperature, precipitation time and pH and speed are considered, although the complete mechanism is not entirely understood from a theoretical point of view. What is know is that even minor changes in the parameters used for the process of making may result in a different outcome. As a result, it is very difficult to simply raise the Sears number while keeping all other properties in the claimed range. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

In other words, based on EP ‘755 there is no suggestion as to how to increase the Sears number to greater than 25 ml/5g and keep the BET surface area from 50 to 700 m<sup>2</sup>/g; the DBP absorption from 100 to 450 g/100 g; the choline chloride absorption from 150 to

400 g/100 g (75% absorption by weight aqueous solution); the CTAB surface area from 50 to 350 m<sup>2</sup>/g; the DBP/choline chloride absorption less than 1.07. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

Contrary to the Examiner's belief, there is no linear relationship between all parameters. If one parameter is changed, it cannot be predicted how the other parameters will change. Therefore, simply increasing the alkaline number in EP '755 does not increase the Sears number while keeping all other parameters constant. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

In fact, the prior art cited by the Examiner discloses that increasing the alkaline number would lead to an increase of the DBP (see Example 19 of Türk). The closest Example of EP '755 (C13) has a Sears number of 22.6 ml/5g, and an alkaline number of 7, DBP = 216 g/100g, CC-absorption = 205 g/100g and DBP/CC = 1,05. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

Türk teaches away from increasing the alkaline number as this would increase the DBP.

Even assuming only a minimal increase of the DBP from 216 g/100g to 220 g/100g, this would cause that the ratio DBP/CC-absorption of example C13 of EP '755 would increase to 1,073 which would be out of the claimed range of the present invention. Türk, however, teaches that the increase of the DBP would be higher than 4 g/100g. Thus, a person of ordinary skill in the art starting with example C13 of EP'755 would not consider increasing the alkaline number because Türk teaches that this would cause a shift of the DBP/CC ratio out of the claimed range. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

Further, the Sears number was increased in the present invention, for example, as disclosed at page 2, lines 17 to 20 of the specification, where it is explicitly pointed out that

the silanol group density (= Sears number) is determined by the alkali number [AN], or in other words by the variation of pH/AN during precipitation. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

The patents cited in EP ‘755 do not sufficiently disclose the variation of pH/AN during the precipitation process. Even considering the complete disclosure of the manufacturing process disclosed in EP ‘755 in combination with the in the references DE 1467019, DE 3144299 and DE 19526476 (cited in EP ‘755), it is found that there is no disclosure by which a Sears number of > 25 could be achieved. This is due to the fact that the Sears number, or in other words the number of silanol groups on the silica surface, is determined by the variation of pH/AN during precipitation, whereas all manufacturing processes disclosed in EP ‘755 in combination with those disclosed in the patents cited therein are incapable of any variation whatsoever of the pH values or alkali numbers. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

Further, the Examiner is referring to similarities of the methods of EP ‘755 and the present invention as indication that similar Sears numbers are achieved. Applicants disagree.

Three different types of precipitated silica, A, B, and C, are described in EP ‘755. All three have different production methods. The methods for the precipitated silica A and B are so-called “pH precipitation methods,” in which the pH is kept constant during the precipitation (see Examples 1 and 2 of EP ‘755). The method of the present invention is a so-called “AN precipitation method,” in other words the alkali number (AN) is kept constant. It was already pointed out in the specification of the present application that pH precipitation methods are something different, and lead to different precipitated silica than AN precipitation methods (see page 3, lines 6-18). Thus, the methods for making precipitated silica types A and B of EP ‘755 are different and cannot result in the precipitated silica of the

present invention. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

The silica C from Example 3 of EP ‘755 is prepared using AN precipitation. The method according to Example 3 of EP ‘755 nevertheless differs decisively from that of the present invention, as the following table shows:

Method parameters	EP ‘755, Example 3	Invention, Examples 1-8
Precipitation time (min)	90	40-65
Precipitation temperature (°C)	91-93	85
Alkali number	7	20-40
Solid content of the suspension (g/l)	72	80-105

As can be seen, numerous significant parameters of the method of the present invention are clearly different. Thus, the precipitation time is not even half as long as in EP ‘755, in some instances. The alkali number is 3-6 times greater. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

The total combination of method parameters brings about the clearly different surface chemistry. Therefore, as shown above, the Sears number is a clear distinction criterion. Sears numbers of greater than 25 are neither disclosed nor made obvious by EP ‘755 even in combination with Türk. See the Rule 132 Declaration of Dr. Lindner signed May 13, 2008 (Exhibit C).

Further, regarding Claims 10 and 17, the specific process steps are not disclosed or suggested in EP ‘755 and Türk, alone or in combination:

**simultaneously metering into an aqueous silicate solution more aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l, wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15**

Or

**simultaneously metering into a vessel an aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l, wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15.**

The Examiner states that EP '755 should not be limited to what is stated in the Examples. See page 4, 2<sup>nd</sup> full paragraph of the Office Action of September 22, 2008. However, there is no disclosure or suggestion anywhere in EP'755 in particular of the claimed precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l. The Examiner cannot just interpret a disclosure into EP'755 that is simply not there. If there is a disclosure of the claimed precipitation period of 40 to 65

minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l, the Examiner is requested to point out where the same is found. In absence of such disclosure, the Examiner has not made a proper *prima facie* case of obviousness because she has not shown all claim limitations in the cited references.

Thus, the rejection of Claims 1, 4-5, 10-12, 14-19 and 22-28 over EP '755 in view of Turk et al should be REVERSED.

**CONCLUSION**

For the above reasons, it is respectfully requested that all the rejections still pending in the Final Office Action be REVERSED.

Respectfully submitted,

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VIII. CLAIMS APPENDIX

1. A precipitated silica having the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g;
DBP/choline chloride absorption	less than 1.07; and
Sears number	greater than 25 ml/5g.

4. The precipitated silica of Claim 1, having a modified Sears number of from at least 25 to 45.

5. The precipitated silica of Claim 1, having a BET surface area of 180-210 m<sup>2</sup>/g, a DBP adsorption of 280-450 g/100g, and a CTAB surface area of 130-200 m<sup>2</sup>/g.

10. A process for preparing precipitated silica, comprising:  
simultaneously metering into an aqueous silicate solution more aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,  
optionally filtering the acidified mixture to obtain a filtered precipitated silica,  
optionally drying the filtered precipitated silica,

wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15, and

wherein said silica has the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g;
DBP/choline chloride absorption	less than 1.07;
Sears number	greater than 25 ml/5g.

11. The process of claim 10, wherein the alkali number is of from 25 to 50.

12. The process of claim 10, further comprising the addition of an electrolyte prior to or during the simultaneous addition of aqueous silicate solution and Lewis and/or Brønsted acid.

14. A method, comprising:

contacting the precipitated silica of claim 1 with a feed additive, a chemical intermediate, or a laundry detergent component.

15. A method, comprising:

contacting the precipitated silica of claim 1 with formic acid, propionic acid, lactic acid, phosphoric acid, choline chloride solution, a plant extract, a melamine resin, a coatings additive, a fragrance, or a detergent.

16. An elastomer, plastic, battery separator, toothpaste, catalyst support or flocculation assistant, comprising:

the precipitated silica of Claim 1.

17. A process for preparing precipitated silica, comprising:

simultaneously metering into a vessel an aqueous silicate solution and a Lewis and/or Brønsted acid over a precipitation period of 40 to 65 minutes followed by reacidifying the mixture to a pH of 7-3.0 to provide to provide an acidified mixture having a solid content of the suspension of from 79.3 to 104 g/l,

optionally filtering the acidified mixture to obtain a filtered precipitated silica,

optionally drying the filtered precipitated silica,

wherein the metered addition of the aqueous silicate solution and the Lewis and/or Brønsted acid is carried out while maintaining a constant alkali number in the mixture of at least 15, and

wherein said silica has the following physicochemical characteristics:

BET surface area	from 50 to 700 m <sup>2</sup> /g;
DBP absorption	from 100 to 450 g/100 g;
Choline chloride absorption	from 150 to 400 g/100 g (75% absorption by weight aqueous solution);
CTAB surface area	from 50 to 350 m <sup>2</sup> /g; and
DBP/choline chloride absorption	less than 1.07;

Sears number greater than 25 ml/5g.

18. The process of claim 17, wherein the alkali number is of from 25 to 50.

19. The process of claim 17, further comprising the addition of an electrolyte prior to or during the simultaneous addition of aqueous silicate solution and Lewis and/or Brønsted acid.

22. The process of Claim 10, wherein said filtering of said acidified mixture is performed.

23. The process of Claim 22, wherein said drying of said filtered precipitated silica is performed.

24. The process of Claim 17, wherein said filtering of said acidified mixture is performed.

25. The process of Claim 24, wherein said drying of said filtered precipitated silica is performed.

26. The precipitated silica of Claim 1, having a modified Sears number greater than 28 ml/5g.

27. The precipitated silica of Claim 1, having a modified Sears number greater than 28 ml/5g.

28. The precipitated silica of Claim 1, having a modified Sears number greater than 28 ml/5g.

IX. EVIDENCE APPENDIX

Exhibit A: Rule 132 Declaration of Dr. Ralph Rausch, signed on April 18, 2007.

Exhibit B: Rule 132 Declaration of Dr. Görl signed October 19, 2007.

Exhibit C: Rule 132 Declaration of Dr. Lindner signed May 13, 2008.

# **Exhibit A**

# COPY

Docket No.: 215150US0

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP: 1754

Gottlieb-Georg LINDNER, et al.

SERIAL NO: 10/079,479

EXAMINER: Nguyen, Ngoc Yen M

FILED: February 22, 2002

FOR: SILICA BY PRECIPITATION AT CONSTANT ALKALI NUMBER, AND ITS  
USE

### DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

Sir: *Dr. Ralf Rausch*

Now comes who deposes and states that:

1. I am a graduate of *chemistry* and received my *ph-* degree in the year *1982*

2. I have been employed by *Degussa* for *19* years as a *chemist* in the field  
of *analytical characterization of silica and head of reference laboratory*  
*for precipitated silica*

3. The following experiments were carried out by me or under my direct supervision  
and control.

The Sears number of an original sample of Product A Versuch 6 of EP '755 (see Table 9 of EP '755) was measured as described on pages 13-15 of the specification and found to be *19,9 ml/5g*. In addition the Sears number of an original sample of Product C Versuch 13 of EP '755 (see Table 9 of EP '755) was measured and found to be *22,6 ml/5g*. Thus the Sears numbers of EP '755 are at least 10% below the Sears numbers as now claimed in Claim 1 of the present invention. Thus, the Examiner's statement that EP '755 has the same Sears number as the present invention is incorrect.

4. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to

be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

5. Further deponent saith not.

*R. Daniel*

Signature

*18. April 2007*

Date

Customer Number

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(OSMMN 05/06)

# **Exhibit B**

# COPY

Docket No.: 215150US0

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP: 1754

Gottlieb-Georg LINDNER, et al.

EXAMINER: Nguyen, Ngoc Yen M

SERIAL NO: 10/079,479

FILED: February 22, 2002

FOR: SILICA BY PRECIPITATION AT CONSTANT ALKALI NUMBER, AND ITS  
USE

### DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Dr. Görl who deposes and states that:

1. I am a graduate of University of Würzburg and received my doctorate degree in the year 1987.
2. I have been employed by Evonik Degussa GmbH for 20 years as a Chemist in different positions in the field of applied technology, marketing and production of precipitated silica. In my current position as Manager of Quality Management / Analytics & Reference Laboratory Silica, I am responsible for test methods, specification, cross-checks, etc. of all the silica plant labs and applied technology labs of Evonik Degussa GmbH worldwide.
3. In the Reference Laboratory Silica which I am heading, the experiments submitted in the Rule 132 Declaration filed May 7, 2007, were performed. The margin of error of this experiment, as performed routinely in our reference laboratory, is +/- 0.25 ml/5g.
4. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section

1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

5. Further deponent saith not.

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Signature

Date

*Ade f/25*  
*October 19 / 2007*

# **Exhibit C**

# COPY

Docket No.:

## IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

GROUP:

SERIAL NO:

EXAMINER:

FILED:

FOR:

### DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Dr. Gottlieb Georg Lindner who deposes and states that:

1. I am a graduate of Marsburg University and received my  
Ph.D. in Chemistry degree in the year 1994.

2. I have been employed by Evonik Degussa GmbH for 10 years as a  
Senior Manager in the field of R&D Silicas.

3. Regarding EP '755, the Examiner has argued that a Sears number of greater than 25 ml/5g is obvious and expected in view of the Sears number of  $22.6 \pm 0.25$  ml/5g of Product C Versuch 13 of EP '755 and in view of  $19.9 \pm 0.25$  ml/5g of Product A Versuch 6 of EP '755.

However, there is a difference of at least 10 % between the claimed Sears number and the Sears number of EP '755. The margin of error is very small ( $\pm 0.25$  ml/5g). This was already discussed in the Rule 132 Declaration of Dr. Ralph Rausch, signed on April 18, 2007 and the Rule 132 Declaration of Dr. Görl signed October 19, 2007 (both of record).

4. The difference of at least 10 % between the claimed Sears number and the Sears number of EP '755 is not obvious. It is very difficult to increase the Sears number and at the same time keep all other parameter as claimed in the ranges as claimed. The Sears number is rather complex in its emergence: factors like temperature, precipitation time and pH and speed

are considered, although the complete mechanism is not entirely understood from a theoretical point of view. What is known is that even minor changes in the parameters used for the process of making may result in a different outcome. As a result, it is very difficult to simply raise the Sears number while keeping all other properties in the claimed range.

5. In other words, based on EP '755 there is no suggestion as to how to increase the Sears number to greater than 25 ml/5g and keep the BET surface area from 50 to 700 m<sup>2</sup>/g; the DBP absorption from 100 to 450 g/100 g; the choline chloride absorption from 150 to 400 g/100 g (75% absorption by weight aqueous solution); the CTAB surface area from 50 to 350 m<sup>2</sup>/g; the DBP/choline chloride absorption less than 1.07.

6. Contrary to the Examiner's belief, there is no linear relationship between all parameters. If one parameter is changed, it cannot be predicted how the other parameters will change. Therefore, simply increasing the alkaline number in EP '755 does not increase the Sears number while keeping all other parameters constant.

7. In fact, the prior art cited by the Examiner discloses that increasing the alkaline number would lead to an increase of the DBP (see Example 19 of Türk). The closest Example of EP '755 (C13) has a Sears number of 22.6 ml/5g, and an alkaline number of 7, DBP = 216 g/100g, CC-absorption = 205 g/100g and DBP/CC = 1,05.

8. Türk teaches away from increasing the alkaline number as this would increase the DBP.

9. Even assuming only a minimal increase of the DBP from 216 g/100g to 220 g/100g, this would cause that the ratio DBP/CC-absorption of example C13 of EP '755 would increase to 1,073 which would be out of the claimed range of the present invention. Türk, however, teaches that the increase of the DBP would be higher than 4 g/100g. Thus, a person of ordinary skill in the art starting with example C13 of EP '755 would not consider increasing the alkaline number because Türk teaches that this would cause a shift of the DBP/CC ratio out of the claimed range.

10. Further, the Sears number was increased in the present invention, for example, as disclosed at page 2, lines 17 to 20 of the specification, where it is explicitly pointed out that the silanol group density (= Sears number) is determined by the alkali number [AN], or in other words by the variation of pH/AN during precipitation.

11. The patents cited in EP ‘755 do not sufficiently disclose the variation of pH/AN during the precipitation process. Even considering the complete disclosure of the manufacturing process disclosed in EP ‘755 in combination with the in the references DE 1467019, DE 3144299 and DE 19526476 (cited in EP ‘755), it is found that there is no disclosure by which a Sears number of > 25 could be achieved. This is due to the fact that the Sears number, or in other words the number of silanol groups on the silica surface, is determined by the variation of pH/AN during precipitation, whereas all manufacturing processes disclosed in EP ‘755 in combination with those disclosed in the patents cited therein are incapable of any variation whatsoever of the pH values or alkali numbers.

12. Further, the Examiner is referring to similarities of the methods of EP ‘755 and the present invention as indication that similar Sears numbers are achieved. Applicants disagree.

Three different types of precipitated silica, A, B, and C, are described in EP ‘755. All three have different production methods. The methods for the precipitated silica A and B are so-called “pH precipitation methods,” in which the pH is kept constant during the precipitation (see Examples 1 and 2 of EP ‘755). The method of the present invention is a so-called “AN precipitation method,” in other words the alkali number (AN) is kept constant. It was already pointed out in the specification of the present application that pH precipitation methods are something different, and lead to different precipitated silica than AN precipitation methods (see page 3, lines 6-18). Thus, the methods for making precipitated silica types A and B of EP ‘755 are different and cannot result in the precipitated silica of the present invention.

The silica C from Example 3 of EP '755 is prepared using AN precipitation. The method according to Example 3 of EP '755 nevertheless differs decisively from that of the present invention, as the following table shows:

Method parameters	EP '755, Example 3	Invention, Examples 1-8
Precipitation time (min)	90	40-65
Precipitation temperature (°C)	91-93	85
Alkali number	7	20-40
Solid content of the suspension (g/l)	72	80-105

As can be seen, numerous significant parameters of the method of the present invention are clearly different. Thus, the precipitation time is not even half as long as in EP '755, in some instances. The alkali number is 3-6 times greater.

The total combination of method parameters brings about the clearly different surface chemistry. Therefore, as shown above, the Sears number is a clear distinction criterion. Sears numbers of greater than 25 are neither disclosed nor made obvious by EP '755 even in combination with Türk.

13. The increase of the Sears number by at least 10 % compared to EP '755 as discussed in item 3 above is significant because it is necessary to achieve a sufficient affinity between the silica of the invention and hydrophilic liquids. Without such affinity the desired DBP/CC ratio of below 1.07 could not be reached. The inventors have found out that – for a given set of other physico-chemical properties of a silica - a borderline exists for the Sears number which has to be exceeded.

14. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false

statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

15. Further deponent saith not.

  
\_\_\_\_\_  
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X. RELATED PROCEEDINGS APPENDIX

None.